Proceedings of the Estonian Academy of Sciences,
2016, **65**, 4, 442–451
doi: 10.3176/proc.2016.4.11
Available online at www.eap.ee/proceedings

ENVIRONMENTAL PHYSICS

Lead-210 in the atmospheric air of North and South Estonia: long-term monitoring and back-trajectory calculations

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Received 7 September 2015, revised 15 February 2016, accepted 16 February 2016, available online 30 November 2016

Abstract. As the first study of long-term atmospheric ^{210}Pb in Estonia, the activity concentration and possible origins of the nuclide were monitored at Tõravere, South Estonia, and Narva-Jõesuu, North-East Estonia, from 2001 to 2008. Activities of ^{210}Pb in weekly collected air filter samples were analysed using HPGe gamma spectrometry. Results show high weekly, seasonal, and yearly variability in the range from 0.12 mBq m^{-3} to 2.76 mBq m^{-3} (median 0.45 mBq m^{-3}) for Tõravere and from 0.08 mBq m^{-3} to 2.53 mB m^{-3} (median 0.43 mBq m^{-3}) for Narva-Jõesuu. No significant correlation between the ^{210}Pb activity concentration and meteorological data was found as far as total weekly precipitation, relative humidity, or air temperature were considered. A strong correlation between the ^{210}Pb concentrations from the two sampling sites was present. Back-trajectory calculations made with the METEX software showed clearly that high ^{210}Pb values in surface air were prevailingly caused by continental air masses from the Eurasian continent, while the low activity concentrations tended to arrive via marine air masses from the North Atlantic. Indications of correlation of atmospheric boundary layer height with ^{210}Pb activity concentration were found.

Key words: Pb-210, back-trajectory calculations, atmospheric air, environmental radioactivity, coast area environment, METEX.

1. INTRODUCTION

It is generally known that our surrounding environment, including the atmosphere, is slightly radioactive. The atmosphere is also an effective environment for the transfer of natural and artificial radionuclides, some of them harmful substances in the air, from place to place. As a relatively clean atmospheric air is obligatory for the life of humans and other biota, numerous atmospheric studies have been carried out, a variety of modelling tools have been developed over the years to learn details about the mobility of particles of different sizes in the atmosphere, etc. (Preiss et al., 1996).

One of the important substances in atmospheric air, regarding both scientific research and everyday air quality, is the naturally occurring radionuclide ^{210}Pb. It is a member of the ^{238}U decay chain, which is naturally present in the Earth's crust. As ^{210}Pb is formed in the chain after the alpha decay of ^{222}Rn, which is an inert gas and thus able to exhale from the soil and to migrate to the atmosphere, transfer in air, and deposit to the ground, a considerable amount of ^{210}Pb is constantly present around us in the environment. Together with other ^{222}Rn progeny ^{210}Pb has a significant impact on the effective dose of ionizing radiation to a human via inhalation and ingestion (see e.g. Vecchi et al., 2005). Unlike the short-lived ^{222}Rn, the progeny ^{210}Pb has a half-life of 22.3 years, with mean residence time in

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the atmosphere about 10 days (varying from 0 days to more than 5 weeks), and is thus a useful tracer for atmospheric movements. It has been noticed that ^{210}Pb atoms tend to attach to the sub-micron sized aerosol particles, which are widely used in atmospheric studies as characteristic parameters (Gaffney and Marley, 2002; Gaffney et al., 2004; Vecchi et al., 2005; Sykora et al., 2007; Paatero et al., 2010; Baskaran, 2011). With its 46.5 keV full energy gamma peak ^{210}Pb is comfortably detectable via non-destructive gamma-ray spectrometry when collected from air on an aerosol filter.

In this study the aerosol particles were collected from the atmospheric air using high-volume air samplers at the Estonian Weather Service's meteorological survey sites at Tõravere (58°15′50″N, 26°27′41″E, South Estonia) and Narva-Jõesuu (59°27′47″N, 28°02′4″E, North-East Estonia). The sampling sites together with the local oil-shale-fired power plants are shown in Fig. 1. Air filters were first analysed via HPGe gamma-ray spectrometry in the laboratory of the Radiation Safety Department, Environmental Board, as part of routine monitoring of atmospheric radioactivity. After that, ^{210}Pb activity concentrations of the same filters have been analysed in the laboratory of environmental physics at the Institute of Physics, University of Tartu, since the monitoring of ^{210}Pb concentration is not part of the Estonian national monitoring programme.

2. MATERIALS AND METHODS

Concentrations of ^{210}Pb activity in weekly collected aerosol filter samples were analysed. The samples from the Tõravere station cover the period from 2001 to March 2008; those from Narva-Jõesuu are from 2001 to May 2008. In total, the current study is based on the analysis results of 359 filter samples for each sampling site.

The aerosol filters used were general GF/A grade glass microfibre filters by Whatman (linear measurements: 28.5 cm × 23.0 cm), whose efficiency for collecting particles of 0.3 µm is considered to be 99.95% or better (GE Healthcare, 2013). The atoms of ^{210}Pb participate in the growth of aerosols with a diameter of 0.07–2 µm, which are known to be the main transporters of pollutants in the atmosphere (Ioannidou et al., 2005; Abe et al., 2010). The filters were exposed to atmospheric air for a week with a help of a high-volume air-sampling device JL-150 HUNTER (Senya OY, Finland) at Tõravere, Tartumaa, South Estonia, and Snow White (Senya OY, Finland) at Narva-Jõesuu, Ida-Virumaa, North-East Estonia. The volume of air pumped through the filter during that time was recorded when removing the filter from the device. The average weekly air volume passing the filter at Tõravere was 23 217 m^3, with a minimum and a maximum of 7 445 m^3 and 27 445 m^3, respectively. At Narva-Jõesuu the average air volume

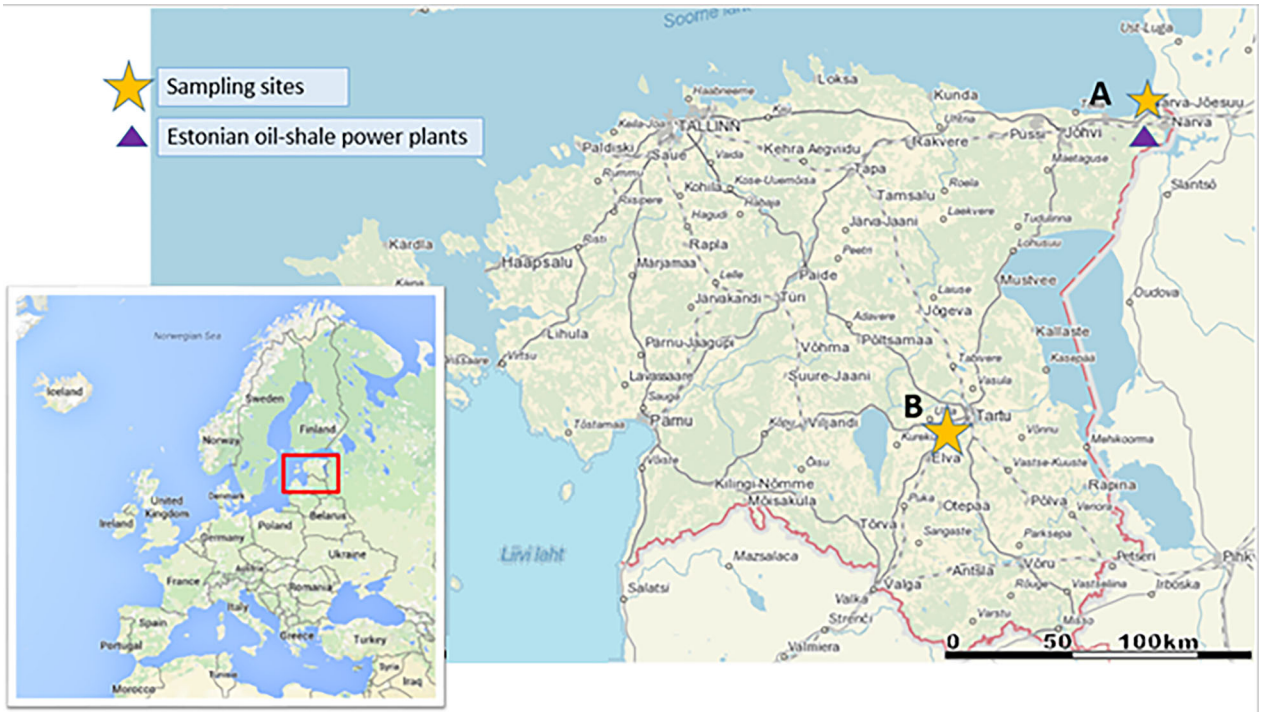

Fig. 1. Map of Estonia. The stars indicate sampling sites: A – Narva-Jõesuu, B – Tõravere. The triangle marks the location of the oil-shale-fired power plants.

was 54 113 m^3, with a minimum of 14 486 m^3 and a maximum of 84 484 m^3. For the calculations of the ^{210}Pb activity concentration the respective weekly values were used.

For analysis the filters were compressed into comparable samples by means of a hydraulic press with special anvils. As a result, every filter became a small cylinder with a diameter of 4.18 cm but of varying height. The samples were placed in the polyethylene beakers of an external diameter of 5.0 cm and height of 2.9 cm. Since the air filters were exposed for a week, the ^{210}Pb activity accumulated in them was enough to determine the activity concentration via 46.5 keV gamma line of ^{210}Pb, although it has emission probability of only 4.25% (LNHB, 2008). Thus no expensive and time-consuming radiochemical analysis is needed.

The activity concentration of ^{210}Pb was measured by the 46.5 keV gamma line using a gamma spectrometer with a planar HPGe detector GPD-50400 (Baltic Scientific Instruments, Latvia) with energy resolution of 610 eV at 59.6 keV. The spectrometer is calibrated using the IAEA RGU-1 reference samples of comparable geometry. Because of the variable sample heights and of relatively low-energy gamma ray, the results were corrected for self-attenuation via efficiency correction factors. In previous studies various theoretical and empirical methods have been used for estimating the self-attenuation coefficients; Monte Carlo simulations were proved to be somewhat time-consuming but the most accurate methods (García-Talavera and Peña, 2004). We used the Geant4 software to create Monte Carlo simulations as a basis for calculating the efficiency correction factors. The method is described in detail in (Isakar et al., 2007). The activity concentration values were decay time-corrected to the middle of the exposure week, the results were an order of magnitude bigger than the estimated lower limit of detection for this method. The results together with combined uncertainties for each measurement are shown in Fig. 2 and Fig. 3.

As the filters were exposed to atmospheric air at the weather station belonging to the Estonian Weather Service, the meteorological data for finding possible correlations were collected comfortably at the same site. The Estonian Weather Service kindly provided the data, including daily averages of temperature, relative humidity, air pressure, and the daily sum of precipitation, from which we found weekly averages or totals, respectively.

For statistical data analysis, including description of ^{210}Pb activity data and correlation tests, the statistical computing environment R together with its graphical user interface RStudio (by RStudio, Inc.) was used, both freeware. In the current study R's 32-bit version i386 2.15.0 and RStudio version 0.98.507, both running on a 64-bit Windows 7 Professional environment (including SP 1), were applied (López-de-Lacalle, 2006; Field et al., 2012). For descriptive statistics and visualization of back-trajectories 32-bit version of Origin Pro 9.0.0 (SR 2) software by OriginLab Corporation (USA) was used.

For back-trajectory calculations 20 weekly samples with the highest and 20 weekly samples with the lowest ^{210}Pb activity concentration data were chosen for both sampling stations. For the Tõravere sampling site this means from 0.12 mBq m^{-3} to 0.21 mBq m^{-3} for the low end and from 1.2 mBq m^{-3} to 2.8 mBq m^{-3} for the high end. For the Narva-Jõesuu sampling site the low values begin with 0.08 mBq m^{-3} and reach 0.20 mBq m^{-3}, and the highest values fall in the region from 1.2 mBq m^{-3} to 2.5 mBq m^{-3}. Air-package transport software METEX

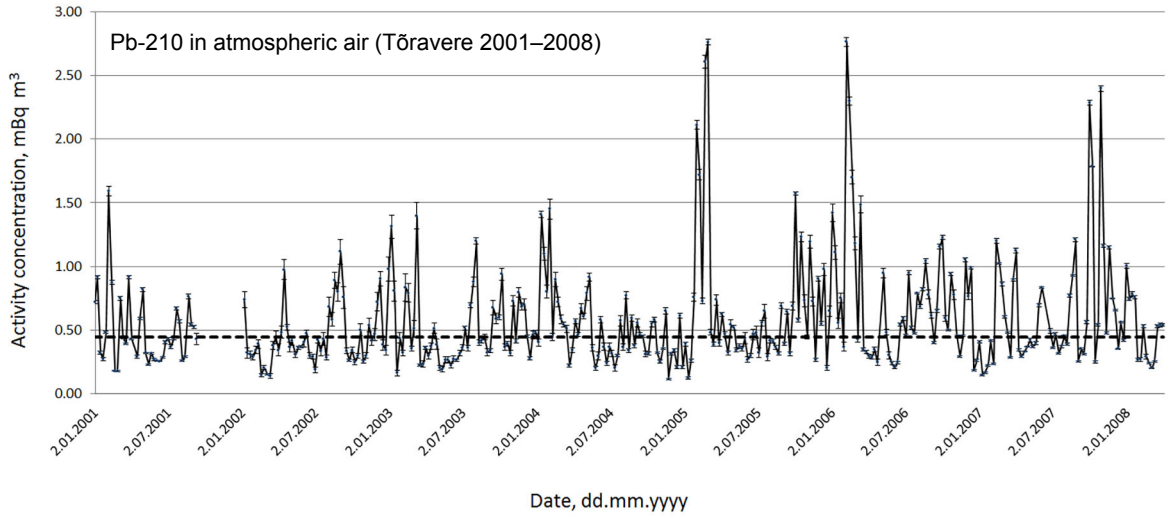

Fig. 2. Concentrations of ^{210}Pb activity in South Estonia from 2001 to 2008 with combined uncertainties. The dashed line indicates the median value of 0.45 mBq m^{-3}.

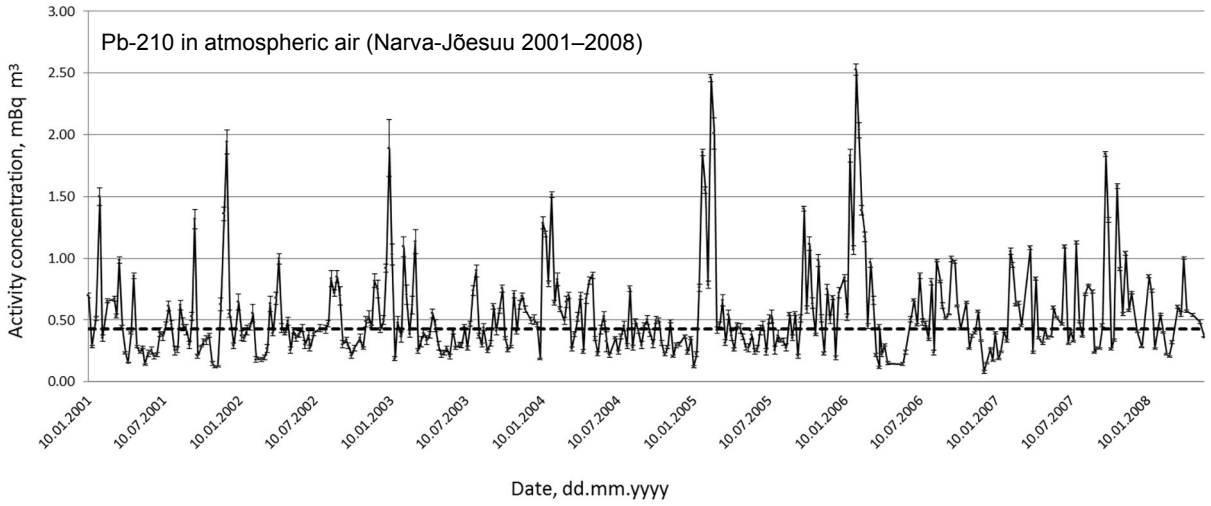

Fig. 3. Concentrations of ^{210}Pb activity in North-East Estonia from 2001 to 2008 with combined uncertainties. The dashed line indicates the median value of 0.43 mBq m^{-3}.

(Zeng et al., 2010) was applied to the respective weeks. For back-trajectory calculations the NCEP (National Centers for Environmental Prediction, USA) Reanalysis Data was used through Simple Object Access Protocol (SOAP), which allows trajectory calculations without downloading a large amount of data to the local computer. The parameters for back-trajectory calculations were optimized considering the air filter exposition time, mean residence time of ^{210}Pb in the atmosphere, and half-life of the parent nuclide. The parameters were kinematic model on the NCEP reanalysis with a trajectory length 96 hours with one calculation after every 6 hours for the whole week when the aerosol filter is exposed to the atmospheric air. The starting moment for calculations was 12:00 on the day when the filter was installed; from this point backward the air-package parameters were found for every 96 hours in the past. The second similar calculation was made starting at 18:00 on the same day, the third starting at 00:00 on the next day, etc. For continuous calculation of 29 trajectories for one week and for compiling the results into one CSV file, two original script files were generated. For the trajectory distribution analysis, considering the origin of air-packages carrying high or low ^{210}Pb concentrations, OriginPro 9 software was used.

3. RESULTS AND DISCUSSION

3.1. Distribution of ^{210}Pb activity concentrations

Over the 2001–2008 sampling period the activity concentration of ^{210}Pb in atmospheric air varied considerably: at Tõravere from 0.12 mBq m^{-3} to 2.76 mBq m^{-3}, with the arithmetic mean of 0.57 mBq m^{-3} and the median of 0.45 mBq m^{-3}; at Narva-Jõesuu from 0.08 mBq m^{-3} to 2.53 mBq m^{-3}, with the arithmetic mean of 0.53 mBq m^{-3} and the median of 0.43 mBq m^{-3}. The measured data over the period are shown in Fig. 2 and Fig. 3. For illustrative purposes the frequency distribution of the Tõravere data is given in Fig. 4. Since the frequency distribution of data is positively skewed (skewnes = 2.53) and with high kurtosis (8.52), the normal distribution was ruled out and for correlation ranks Kendall's tau was introduced (Kowalski, 1972; Newson, 2002; Fredricks and Nelsen, 2004). As seen in Fig. 4, the frequency

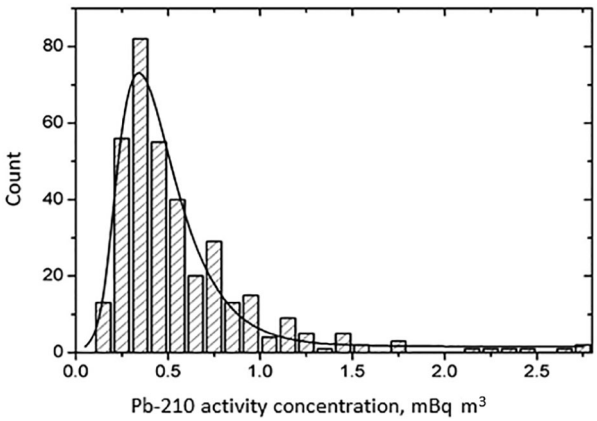

Fig. 4. Distribution of ^{210}Pb activity concentration data for Tõravere, South Estonia. The data is positively skewed (skewness = 2.53) and has a high kurtosis (8.52). The solid line is a least square fit assuming log-normal distribution (see the text).

distribution can be fitted reasonably well ($R^2 = 0.96$) by means of the log-normal distribution:

$$y = y_0 + \frac{A}{\sqrt{2\pi}\cdot w\cdot x}\cdot \exp\left(-\frac{\left(\ln\frac{x}{xc}\right)^2}{2\cdot w^2}\right), \quad (1)$$

where $xc = 0.42 \pm 0.01$, $w = 0.46 \pm 0.02$, $A = 31.1 \pm 1.4$. The parameters denoted *xc* and *w* are, respectively, the mean and standard deviation of the variable's natural logarithm. The frequency distribution for the Narva-Jõesuu data follows practically the same pattern.

According to Preiss et al. (1996), both Tõravere and Narva-Jõesuu, Estonia, lie in the category of the Continental Area, where the mean surface air concentrations of ^{210}Pb are about 0.53 mBq m^{-3}, although they acknowledge significant differences in the average values depending on the particular sampling site, since the influence of continental and marine air masses varies in a wide range. For example, over the years 1985 to 1996 in Belgrade, Serbia (44°47′N, 20°32′E), ^{210}Pb concentrations varied from 0.30 to 3.17 mBq m^{-3} (Todorovic et al., 2000). From 1995 to 1996 in Palermo, Italy (38°07′N, 13°22′E), concentrations from 0.14 to 3.39 mBq m^{-3} were reported (Cannizzaro et al., 1999). In 2000–2001 in Milan, Italy (45°26′N, 09°17′E), the variation of the activity concentrations of ^{210}Pb was from 0.17 to 2.20 mBq m^{-3} (Vecchi et al., 2005). In 2000–2003 in Bratislava, Slovakia (48°09′N, 17°06′E), ^{210}Pb activity concentrations from 0.27 to 3.07 mBq m^{-3} were measured (Sykora et al., 2007). Long-term monitoring of ^{210}Pb in Thessaloniki, Greece (40°38′N, 22°58′E), over the years 1987–2001 showed results from 0.11 to 1.98 mBq m^{-3} (Ioannidou et al., 2005).

Results from Tõravere and Narva-Jõesuu are thus in a good agreement with similar studies around Europe both with regard to the mean values of activity concentration and the wide variability of data. A closer look at the data from Tõravere and Narva-Jõesuu and comparison with the results for Harku (Tallinn), North Estonia (59°24′N, 24°36′E), over the years 2001–2005 (Realo et al., 2007), show that at Harku the data variation from 0.07 to 2.02 mBq m^{-3} is slightly less than at Tõravere and Narva-Jõesuu. At the same time, in general, a good agreement is observed for all sites regarding the frequency distribution of activity concentrations and the tendency of measuring higher values in wintertime. The concentrations of ^{210}Pb measured in all three sampling stations, Harku, Tõravere, and Narva-Jõesuu, correlate with each other pretty well. Harku data correlates with Tõravere and Narva-Jõesuu with Kendall's tau value 0.57 and 0.59, respectively, while Tõravere and Narva-Jõesuu correlate with each other with Kendall's tau equal to 0.68. In all cases the *p*-value is well below the R's limit 2.2×10^{-16}. Correlation of the latter two, especially of high ^{210}Pb concentrations, is also visible while comparing Fig. 2 and Fig. 3.

3.2. Seasonal variations

For seasonal analysis the seasons are defined according to the meteorological situation in Estonia: winter consists of December, January, and February; spring lasts during March, April, and May; summer months are June, July, and August; and autumn lasts from September to November.

High ^{210}Pb activity concentrations in winter, closely followed by autumn data, are demonstrated in Fig. 5 and Fig. 6. The latter presents the ^{210}Pb data as a modified box and whiskers plot. The dark horizontal line in the middle of the box indicates the median value, bottom and top lines for constructing the box are equal to the 1st and 3rd quartile of data; thus 50% of data lie 'in the box'. Whiskers are calculated here as (3rd *quartile* + *IQR* · 1.5) and (1st *quartile* – *IQR* · 1.5), where *IQR* is the difference of the 1st and the 3rd quartile. Data outside the reach of whiskers are called outliers. It can be seen from Fig. 5 and Fig. 6 that in case of ^{210}Pb outliers occur as abnormally high values only and are never lower than the 1st quartile values; this follows also from the positive skewness of data distributions (Fig. 4).

The same seasonal tendency has been detected previously in a number of sampling stations all over Europe (Paatero et al., 1998, 2010; Arimoto et al., 1999; Todorovic et al., 2000; Winkler and Rosner, 2000; Dueñas et al., 2004; Ioannidou et al., 2005; Realo et al., 2007; Abe et al., 2010). Reasons for seasonal variation of ^{210}Pb concentrations in the air are not yet fully clear. Baskaran (2011) explained a contradicting phenomenon, namely the lower concentrations of ^{210}Pb in higher latitudes (60° to 75°N) in winter, by decreased radon emanation from the ground due to the snow cover and high humidity in the soil. Paatero et al. (1998), on the contrary, suggested that Rn emanation rates and ^{210}Pb concentrations should not be directly linked since ^{210}Pb in air is highly influenced by wet deposition while Rn is not. Later, based on earlier studies and literature data, they suggested that the reasons for high activity concentrations during winter might be low levels of precipitation, reduced air chemistry, and stagnant mixing conditions, as these factors increase the aerosol residence time, which facilitates the accumulation of ^{210}Pb in air (Paatero et al., 2010). We have found indications for the latter: analysis of the atmospheric boundary layer data from back-trajectory calculations suggested that the ^{210}Pb concentration was linked to the mean and median boundary layer heights of the lifetime of the modelled trajectory, described below. In addition, we cannot ignore the influence of local meteorological parameters: atmospheric pressure, mixing layer height, temperature changes, humidity of the soil, and possible snow cover. The prevailing assumption so far is that higher values in winter and lower values in summer are caused respectively by relatively passive or active vertical mixing of air masses in the atmosphere.

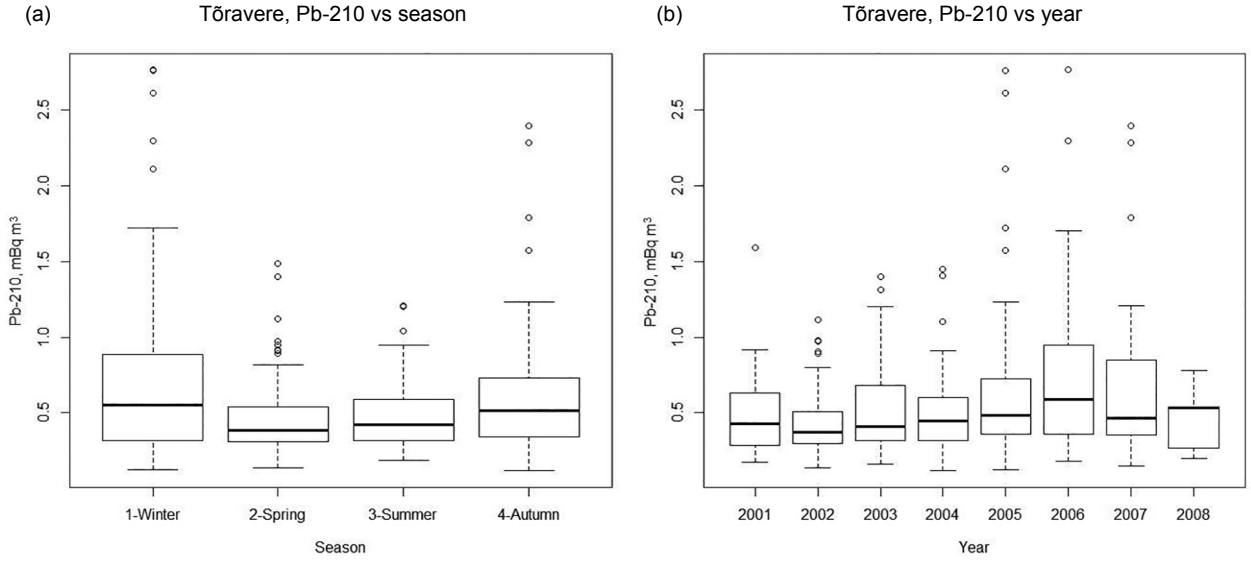

Fig. 5. Variation of the ^{210}Pb activity concentration at Tõravere, South Estonia, over the seasons (a) and years (b). Bold lines indicate the median values, which are surrounded by interquartile range; whiskers mark the one and a half times the interquartile range. Points lying separately are considered outliers and are a result of non-normally distributed data (see Fig. 4, detailed description in text).

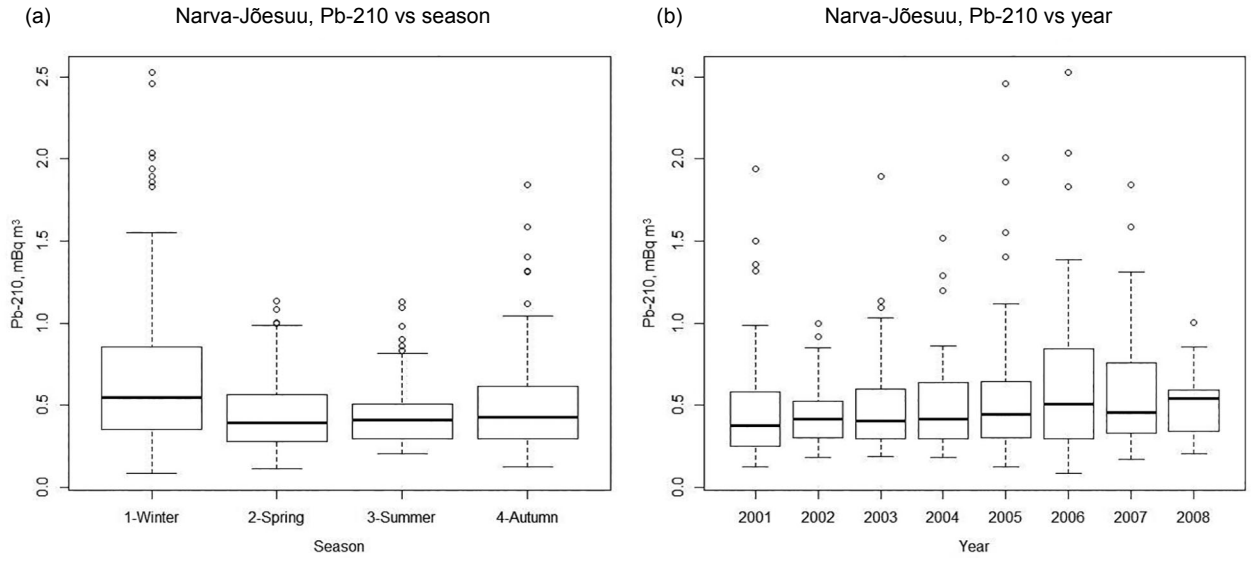

Fig. 6. Variation of the ^{210}Pb activity concentration at Narva-Jõesuu, North-East Estonia, over the seasons (a) and years (b). Bold lines indicate the median values, which are surrounded by interquartile range; whiskers mark the one and a half times the interquartile range. Points lying separately are considered outliers and are a result of non-normally distributed data (see Fig. 4, detailed description in text).

The latter occurs in case of warmer weather, thus in summer (Paatero et al., 1998).

We tried to take those factors into account and find correlations of ^{210}Pb activity concentration with local meteorological data. Because of the wet deposition effect, a relatively strong correlation between precipitation and the activity concentration of ^{210}Pb in surface air might be expected. Nevertheless, while considering the annual or weekly average concentrations of ^{210}Pb and total precipitation, no evidence of reasonably strong correlation has been detected either in our previous studies (Realo et al., 2007) or in the current analysis. The possible reason is the influence of the other factors mentioned earlier (Preiss et al., 1996; Arimoto et al., 1999; Ioannidou et al., 2005). Results of correlation tests revealed only a slight positive correlation of the ^{210}Pb activity concen-

Table 1. Correlation of ^{210}Pb activity concentrations with meteorological data obtained from the same sampling site. τ – correlation coefficient; p and z – test values to identify statistical significance of τ

Meteorological data	Tõravere			Narva-Jõesuu		
	z	p	τ	z	p	τ
Atmospheric pressure	6.8	9.8×10^{-12}	0.26	7.4	1.3×10^{-13}	0.26
Relative humidity	0.35	0.73	0.013	1.4	0.15	0.051
Temperature	–0.35	0.73	–0.013	–2.8	0.0058	–0.098
Precipitation	–4.4	9.2×10^{-6}	–0.17	–6.6	5.0×10^{-11}	–0.23

tration with atmospheric pressure and an even weaker negative correlation with precipitation (Table 1). Kendall's rank correlation was tested via p-value and z-value. Small p-values together with large z-values indicate the correlation with atmospheric pressure and precipitation to be statistically significant, while the nearly non-existent correlation of ^{210}Pb concentrations with humidity and temperature might well be a coincidence.

3.3. Back-trajectory calculations

In general, the main processes influencing the concentration of ^{210}Pb in air are (1) local sources emanating Rn to the air and (2) transport (and deposition) of atmospheric aerosol particles carrying ^{210}Pb via atmospheric circulation and precipitation. Local sources mean in most cases, including Tõravere and Narva-Jõesuu, ^{222}Rn exhalation from the local earth crust (Preiss et al., 1996). The Narva-Jõesuu sampling site (Fig. 1) is located near two large oil-shale-fired power plants, the Estonian Power Plant and the Baltic Power Plant, about 11 km and 18 km south/southwest, respectively. Both can be considered as local ^{210}Pb sources (Realo et al., 1996; Vaasma et al., 2014a, 2014b).

Considering the strong correlation between the ^{210}Pb concentrations at Tõravere and Narva-Jõesuu, as previously discussed, it can be assumed that the influence of local sources, e.g., power plants, is negligible compared to that of long-distance air masses. The same conclusion follows from negative results of the performed correlation analysis of ^{210}Pb concentrations with meteorological data. On the other hand, a week may be too long integration time; thus further studies with shorter exposition times might be needed for more detailed conclusions. This regards especially human activities near the Narva-Jõesuu site.

To get an overview of the features of long-range transport of ^{210}Pb, the back-trajectory calculation method with the use of global meteorological data for the description of air parcels in the atmosphere was applied.

The geographical distribution of back-trajectories originating at Tõravere and Narva-Jõesuu is shown in Fig. 7 and Fig. 8, respectively. Each picture consists of 50 440 air parcel hourly coordinates corresponding to 20 weeks under the detailed study. Each figure presents a contour plot of the result of OriginPro 9 descriptive statistical analysis as a 2D histogram, where numbers of air parcel trajectory hourly coordinates are summed in every 2D bin of 1°E × 0.2°N. For contour plotting the bin sums were subdivided into nine levels in logarithmic scale. The geographical distribution depends on the number of trajectories travelling through the specific bin as well as on the velocity of any air parcel.

The origin of air parcels arriving at Tõravere and Narva-Jõesuu sampling sites in the weeks characterized by high ^{210}Pb concentrations is shown in Fig. 7a and Fig. 8a, respectively. In both cases the higher number of air parcel coordinates originate from southeast of the sampling site, thus from the Eurasian continental region. The origin of air parcels arriving at the same sampling sites in the weeks when low ^{210}Pb concentrations were measured is shown in Fig. 7b and Fig. 8b, respectively. The variability of origin and distribution of back-trajectories is much larger than in the case of high ^{210}Pb concentrations. There is a tendency that low ^{210}Pb concentrations are mainly related to marine air masses from the west, with a slight addition from south-west and north-west. This confirms the hypothesis connecting high ^{210}Pb activity concentrations in the Estonian air to the exhalation of ^{222}Rn from the ground of the Eurasian continent and its long-range transport, although probably the link is much more complicated. It has been shown in earlier studies that transfers of ^{222}Rn and ^{210}Pb are not directly linked because of their different behaviour with wet deposition (Paatero et al., 1998, 2001, 2010).

After analysing the height distribution of air parcel trajectories under study, we found it reasonable to assume that the vertical movements included in trajectories follow such complex patterns that in case of our time scale the mean and median height values for air parcels carrying minimum and maximum ^{210}Pb activity concentrations are almost the same. For the Tõravere sampling site the mean height of air parcels over 20 weeks was 787 m above sea level in the case of minimum ^{210}Pb activities and 738 m in the case of maximum activities. In Narva-Jõesuu the mean heights were 753 m and 749 m, respectively, for minimum and maximum ^{210}Pb activities.

The NCEP data include information about the height of the atmospheric boundary layer for every air parcel coordinate. Here might lie an important explanation for

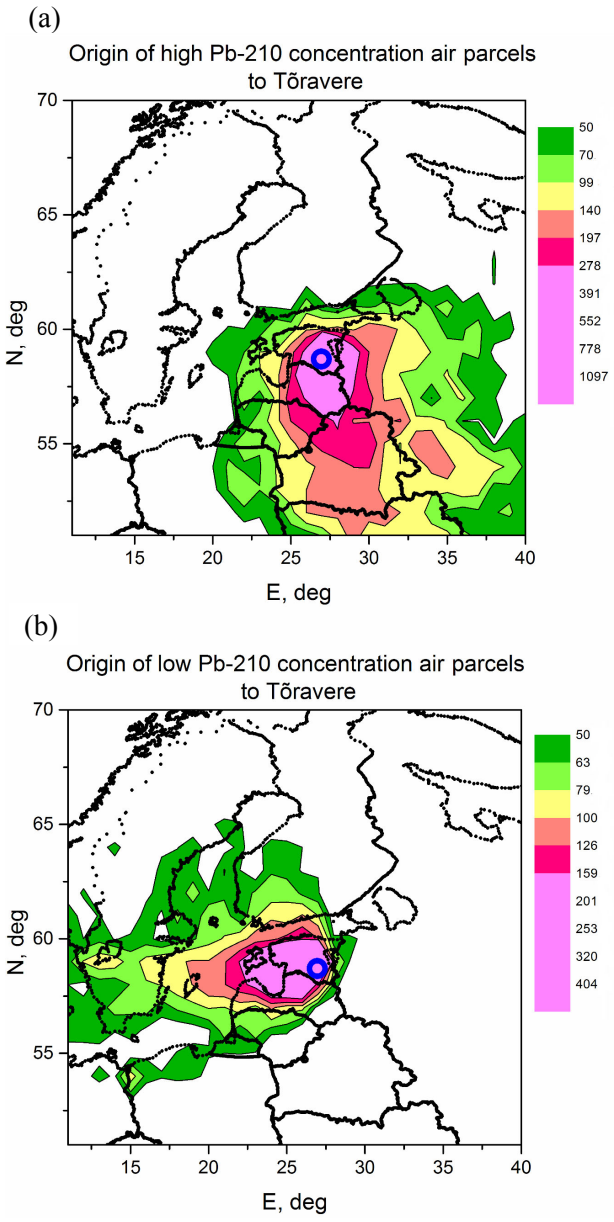

Fig. 7. Origin of air parcels arriving at Tõravere, South Estonia, carrying high (a) and low (b) ^{210}Pb concentrations. Each picture describes 20 weeks with the highest or the lowest ^{210}Pb concentrations. Levels indicated by different colours are in logarithmic scale. The blue circle marks the sampling site.

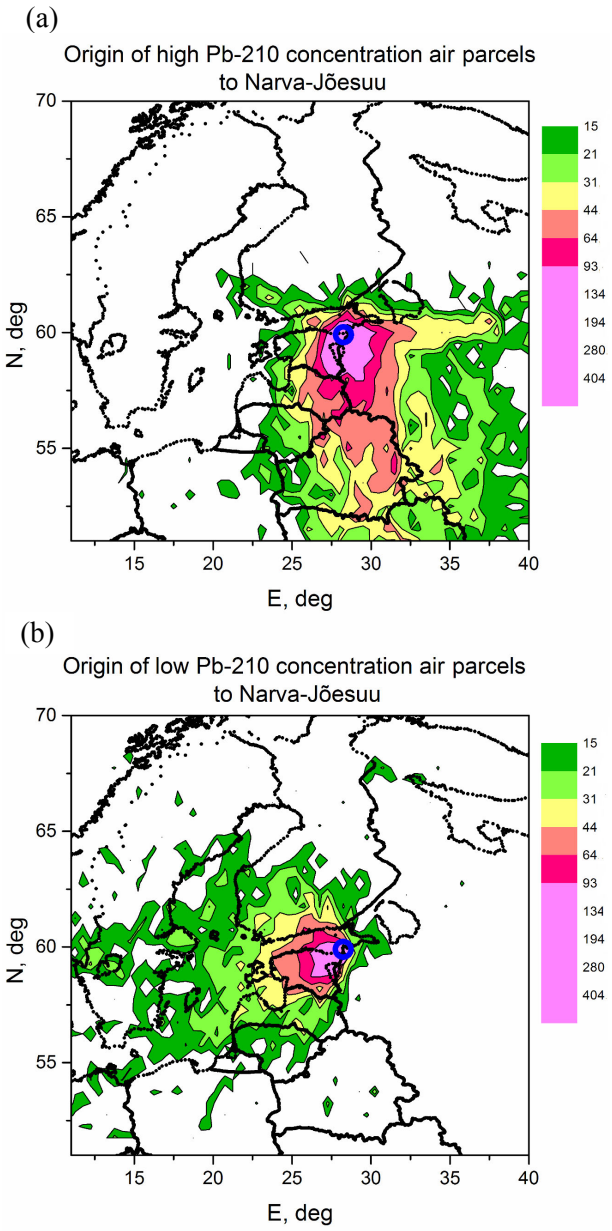

Fig. 8. Origin of air parcels arriving at Narva-Jõesuu, North-East Estonia, carrying high (a) and low (b) ^{210}Pb concentrations. Each picture describes 20 weeks with the highest or the lowest ^{210}Pb concentrations. Levels indicated by different colours are in the logarithmic scale. The blue circle marks the sampling site.

the seasonal variation of ^{210}Pb activity concentrations. Detailed study of boundary layer heights calculated for trajectories showed that in the case of a mean height of 777 m at Tõravere and 711 m at Narva-Jõesuu, low ^{210}Pb activity concentrations were measured. High ^{210}Pb concentrations were measured when boundary layer heights were much lower, e.g., only 418 m at Tõravere and 411 m at Narva-Jõesuu. Again, as pointed out earlier, shorter exposition times of air filters and further analysis might be needed to verify these results.

Annual variation in the activity concentration of ^{210}Pb (Figs 5 and 6) is often explained with the zonal circulation in the atmosphere, which tends to move air masses in the latitudes from 30°N to 60°N from west to east and leads to concentration differences in the western and eastern coasts of the continents. Depending on the origin of air masses, the surface air becomes enriched with or depleted of ^{210}Pb (Preiss et al., 1996). At Tõravere

and Narva-Jõesuu, which lie near the western coast of the Eurasian continent, we might assume lower annual mean concentrations of ^{210}Pb than at its eastern coast due to the influence of the marine, low ^{210}Pb, air masses from the North Atlantic Ocean. However, Suzuki and Shiono (1995) reported that in 1992 the measured ^{210}Pb activity concentrations varied from 0.07 to 1.00 mBq m^{-3} in Honshu Island, Japan (38°46′N, 139°44′E); these values are clearly lower than the concentrations in any sampling site in Estonia a decade later. In cases like this it is reasonable to suggest that continental air masses from the Eurasian continent prevail over marine air masses at the Tõravere and Narva-Jõesuu sites and vice versa at the Honshu site. The huge difference between low and high values of ^{210}Pb activity concentrations measured at Tõravere and Narva-Jõesuu together with back-trajectory calculation results leave no doubt about the importance of the origin of air masses.

4. CONCLUSIONS

We measured activity concentrations of ^{210}Pb from January 2001 to March 2008 at Tõravere, South Estonia, and from January 2001 to May 2008 at Narva-Jõesuu, North-East Estonia. Our results show high weekly, seasonal, and yearly variability, where low and high activity concentrations differ by an order of magnitude. The general tendency was to observe higher concentration values of ^{210}Pb and their wider variability in winter, and lower concentrations and smaller variability in summer. Since no clear correlation with meteorological data was found, but a high positive correlation between the sampling sites was present, we explain the summer–winter difference with the different origin of air masses and the domination of the influence of long-range transport over local sources. Back-trajectory calculations conducted with the METEX software suggest that high ^{210}Pb values in the surface air at Tõravere and Narva-Jõesuu are caused by continental air masses transported from the Eurasian continent, while the low activity concentrations tend to arrive via marine air masses from the North Atlantic Ocean. In addition, a possible influence of the height of the atmospheric boundary layer was detected, suggesting that further studies are needed.

ACKNOWLEDGEMENTS

We thank colleagues from the Radiation Safety Department, Environmental Board, Tallinn, especially Eia Jakobson, for long-term cooperation and presenting the air filters for reanalysis. We thank the Estonian Weather Service for providing us with the meteorological data needed. We are also grateful to Eur Ing Enn Kukk for help in making back-trajectory calculations more time-efficient. This work was partially supported by the European Union via the Regional Development Fund (project No. 3.2.0801.11-0033). The publication costs of this article were covered by the Estonian Academy of Sciences.

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^{210}Pb Põhja- ja Lõuna-Eesti atmosfääriõhus: pikaajaline seire ning päritolu hindamine

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On kajastatud esmakordselt pikaajalise atmosfäärse ^{210}Pb seire tulemusi koos ^{210}Pb päritolu hinnanguga. ^{210}Pb proove koguti filterseadmete abil Lõuna-Eestis Tõraveres ja Põhja-Eestis Narva-Jõesuus aastail 2001–2008. Iga kogumistsükkel kestis nädala ja filtri hilisemal gammaspektromeetrilisel analüüsil määrati nädala kumulatiivne ^{210}Pb aktiivsuskontsentratsioon. ^{210}Pb kontsentratsioonid varieerusid nädalate, aastaaegade ja aastate lõikes märkimisväärselt: 0,12 mBq m^{-3} kuni 2,76 mBq m^{-3} Tõraveres (mediaan 0,45 mBq m^{-3}) ning 0,08 mBq m^{-3} kuni 2,53 mB m^{-3} Narva-Jõesuus (mediaan 0,43 mBq m^{-3}). Kõrgemad väärtused ja suurem varieeruvus ilmnesid talviti, madalad aktiivsuskontsentratsioonid ning nende väiksem varieeruvus iseloomustasid suveperioode. Tarkvarapaketi METEX abil hinnati kõrge ja madala ^{210}Pb kontsentratsiooniga õhupakettide päritolu. Nn tagasitrajektooride (*back-trajectories*) arvutused näitasid, et suure ^{210}Pb kontsentratsiooniga õhk saabub nii Narva-Jõesuusse kui ka Tõraverre kontinentaalsete õhumassidega Euraasiast, samas kui madala ^{210}Pb kontsentratsiooniga õhk on merelist päritolu, saabudes meile Põhja-Atlandilt.

Korrelatsiooni ilmastikunähtuste (temperatuur, suhteline õhuniiskus, sademed) ja ^{210}Pb kontsentratsiooni vahel ei leitud. Küll aga tuvastati nõrk korrelatsioon ^{210}Pb kontsentratsiooni ja atmosfäärse piirkihi kõrguse vahel. Viimane nõuab autorite hinnangul veel edasisi uuringuid.

Tõraveres ja Narva-Jõesuus mõõdetud ^{210}Pb kontsentratsioonide vahel leiti väga tugev positiivne korrelatsioon. Sellest järeldavad autorid, et kaugemat päritolu õhumasside mõju ^{210}Pb kontsentratsioonile on suurem kui lokaalsete allikate, sh tööstus, oma.